SOFCs Based on $SrCe_{0.95}Yb_{0.05}O_3$ Proton Conductor

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INTRODUCTION

$SrCe_{0.95}Yb_{0.05}O_3$ perovskite is a solid electrolyte exhibiting high proton conductivity between 600-1000°C [Iwahara et al, *Solid State Ionics* 3-4 (1981) 359]. We have fabricated a simple self-supported solid oxide fuel cell (SOFC) based on this material with Pt electrodes. Experiments have been carried out under H_2 and CH_4 fuels to evaluate fuel cell performance. In particular we were interested in quantifying the additional electrical resistance associated with the operation of the fuel cell under the internal reforming/decomposition of CH_4 fuel.

EXPERIMENTAL

$SrCe_{0.95}Yb_{0.05}O_3$ powder was prepared by Praxair Speciality Ceramics. 1 mm thick pellets were formed and sintered at 1450°C to > 95% density. Thin (ca. 100 nm) Pt electrodes were applied to either side of the pellet by magnetron sputtering and current collection was enhanced by the application of Englehard Pt paint on top of the sputtered electrodes. An external potentiostat/galvanostat was used to control the current output and measure the voltage in d/c fuel cell tests. H_2 or CH_4 fuel was fed to the anode while the cathode was exposed to air. The fuel was fed dry or saturated with water at room temperature.

RESULTS

Initial fuel cell experiments were carried out at 600-1000°C with humidified H_2 fuel at the anode and air at the cathode. The theoretical EMF of the cell can be calculated from the Nernst equation to be approximately 1.1 V. The experimental open circuit voltage (OCV) was approximately consistent with the theoretical at 600°C. But approximately 0.1 V was lost at 800°C and the same again at 1000°C. This indicated some loss of efficiency due to the introduction of electronic conductivity in the electrolyte at higher temperature.
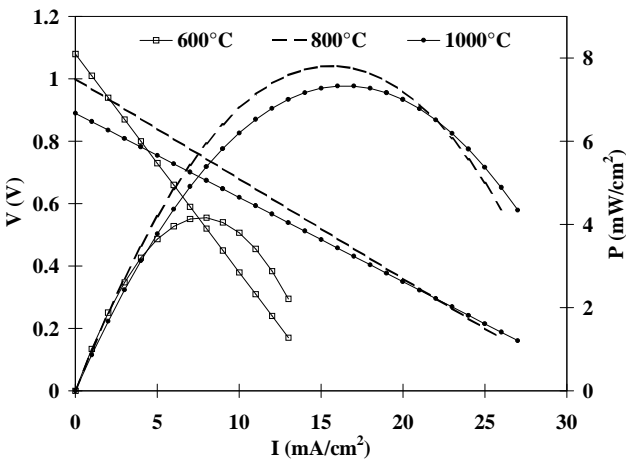

Figure 1. Fuel cell performance under saturated H_2

The linear current-voltage relationships shown in Figure 1 allow an ohmic gradient to be extracted at each temperature. The corresponding area specific resistance (ASR) was much lower at 800°C compared to 600°C as a result of the increased electrolyte conductivity and electrode kinetics. However at 1000°C versus 800°C the ASR was only slightly lower; this is because the conductivity of electrolytes like $SrCe_{0.95}Yb_{0.05}O_3$ tends to decrease at high temperatures [Norby, *Solid State Ionics* 125 (1999) 359].

The maximum power density (P_{max}) of the fuel cell under humidified H_2 was nearly 8 mW/cm^2 at 800°C, approximately double that at 600°C (see Figure 1). But there was no advantage at 1000°C for the reasons outlined above. We note that the fuel cell performance, for example the ASR at 800°C, was a factor of 3-4 worse than the expected performance given the known protonic conductivity of $SrCe_{0.95}Yb_{0.05}O_3$ (0.006 S/cm at 800°C) [Iwahara, in: *Proton Conductors*, Cambridge University Press, 1992, p. 511]. This may be due to significant impedance associated with the electrodes.

Focussing on the results at 800°C in Table 1, it can be seen that the ASR across the fuel cell was approximately the same under dry and saturated H_2 fuel. However the ASR for the case of saturated CH_4 was somewhat higher. This shows that there is additional impedance associated with the internal reforming of CH_4 to H_2 at the anode. The reforming reaction in the presence of water (~ 3% H_2O would be expected from the room temperature saturation) is as follows (see reaction below). Product analysis by mass spectrometry was consistent with this reaction – a significant proportion of the anode effluent contained CO and H_2. The voltage readings in this case showed oscillatory output which may be attributed to complex kinetic behaviour as a result of CO interaction with the Pt catalyst [Turner and Maple, *Surface Science* 147 (1984) 647].

$$CH_4 + H_2O \rightarrow CO + 3H_2$$
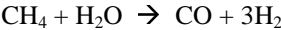
On the other hand, in dry CH_4 there was no oscillation but the ASR was significantly higher. This gives an indication that the generation of H_2 at the anode was much slower, leading to a much higher impedance at the anode. In this case H_2 would be produced by the decomposition of CH_4 (as shown in the reaction below). In accordance with this, carbon deposition was observed visually after the experimental apparatus was dismantled.

$$CH_4 \rightarrow C + 2H_2$$

The results show that the fuel cell can be operated under direct CH_4 fuel, but it is important to humidify the fuel to avoid CH_4 decomposition and carbon deposition.

Table 1. Fuel cell results with different fuels at 800°C

Fuel	$H_{2(sat)}$	$H_{2(dry)}$	$CH_{4(sat)}$	$CH_{4(dry)}$
OCV (V)	1.00	1.01	0.98*	1.02
ASR ($\Omega.cm^2$)	32	31	37	58
P_{max} (mW/cm^2)	7.8	8.2	6.5	4.5

* upper limit of oscillating electrical output